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TITLE: Chemical reaction vessel - contains channel coil and thermal probes to control temperature

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BASIC-ABSTRACT:

An insulated chemical reactor comprises: (a) a reaction vessel (110) having inner and outer surfaces; (b) an evacuated insulation shell (300); (c) a temperature controlling helical channel coil (100) outside of the reaction vessel and within the shell. The reaction vessel wall (120) forming one wall of the channel coil. Also claimed are the apparatus including a thermal probe for cooling the contents of a reaction vessel, apparatus with a number of probes and apparatus with gas discharged from the probe being supplied to the coil.

~~USE - Used in controlling the temperature within a chemical reactor.~~

ADVANTAGE - The channel coil is shaped to have two straight, parallel sides of the coil in contact with the reactor normal to the surface

of the outside wall of the reactor. This right angle contact between the channel coil and reactor wall increases the section modulus of the vessel wall and so increase the mechanical strength of the reactor wall. The wall can thus be made thinner to promote better heat transfer.across the wall. The utilisation of an evacuated shell results in greater temperature control of the reaction vessel contents. The heating and boiling of the liquid introduced into the thermal probe provides for the removal of heat from the reactor contents. By maximising turbulent flow immediately adjacent to the probes, by use of an agitator, the thickness of the laminar thin film at the surface of the probe may be minimised. Minimising the thickness of this film is important in preventing material from solidifying, forming ice, on the surface of the probe and s o red

CHOSEN-DRAWING: Dwg.5/7

TITLE-TERMS: CHEMICAL REACT VESSEL CONTAIN CHANNEL COIL THERMAL PROBE CONTROL TEMPERATURE

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TITLE: TEMPERATURE CONTROLLED REACTION VESSEL
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ABSTRACT:

CHG DATE=19990905 STATUS=C>A novel reaction vessel apparatus is provided. The reaction vessel includes internally placed temperature controlling thermal probes in which liquid is boiled. The energy of vaporization is supplied by the reaction vessel contents. The vapor produced by the boiling may be directed to channel coils which surround the outside of the reaction vessel wall. The channel coils contact the outside wall of the reaction vessel perpendicularly, and provide mechanical support for the reaction vessel. The mechanical support from the channel coils allows for a decrease in the thickness of the reaction vessel wall and corresponding increased heat transfer efficiency between the channel coil contents and the reaction vessel contents. The entire above described apparatus is enclosed within an evacuated shell which provides additional insulation.

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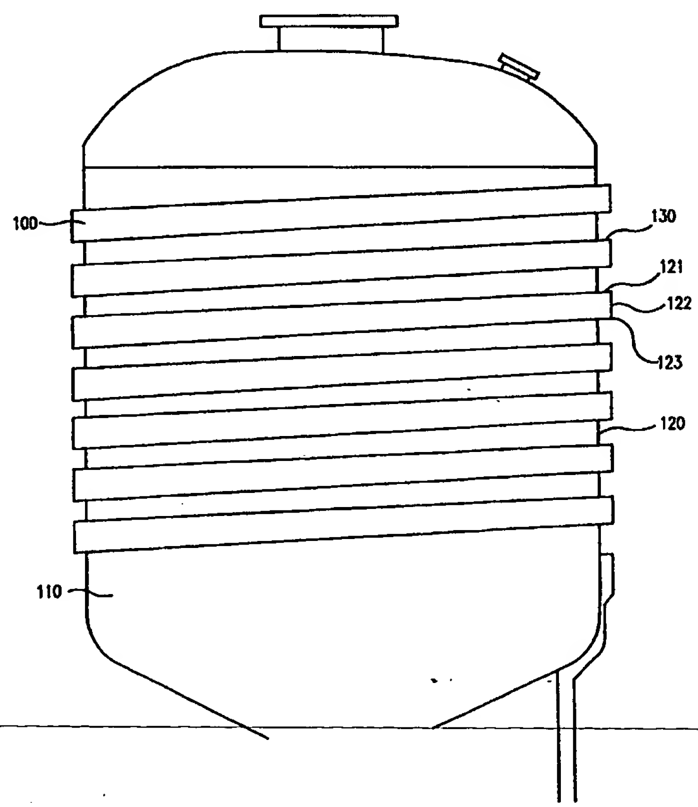
(54) Title: TEMPERATURE CONTROLLED REACTION VESSEL

(57) Abstract

A novel reaction vessel apparatus is provided. The reaction vessel includes internally placed temperature controlling thermal probes in which liquid is boiled. The energy of vaporization is supplied by the reaction vessel contents. The vapor produced by the boiling may be directed to channel coils which surround the outside of the reaction vessel wall. The channel coils contact the outside wall of the reaction vessel perpendicularly, and provide mechanical support for the reaction vessel. The mechanical support from the channel coils allows for a decrease in the thickness of the reaction vessel wall and corresponding increased heat transfer efficiency between the channel coil contents and the reaction vessel contents. The entire above described apparatus is enclosed within an evacuated shell which provides additional insulation.



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TEMPERATURE CONTROLLED REACTION VESSEL

FIELD OF THE INVENTION

The present invention relates to chemical or biological reactors generally, and in particular to an apparatus for controlling the temperature within a chemical reactor.

BACKGROUND OF THE INVENTION

Temperature control of a chemical reaction is often the key to obtaining desired products. Where the temperature is controlled, generally the reaction kinetics are controlled. Where the kinetics are controlled, undesired intermediates and byproducts can be diminished or avoided. Traditional temperature control of industrial reactors is generally attained in one of two ways. One method is to control the temperature of the reactants as they enter the reactor. This method fails to address the heat of reaction, which is often responsible for the majority of heat produced or absorbed in a reaction. The heat of reaction can then alter the temperature of the reactants to produce undesirable products. This is especially true for tank reactors.

Conversely, endothermic reactions require the addition of heat during the reaction to maintain the temperature of the reactants. Again, preadjustment of the temperature of reactants fails to adequately address this situation. Further, complicated production processes may have exothermic and endothermic reactions taking place (usually at different times) as reactants are added or products withdrawn. Preadjustment of reactant temperature is clearly totally inadequate in such situations.

A second method of temperature control of industrial reactors involves the placement of a jacket around the outside of the reaction vessel. In such a case, a fluid of desired temperature is passed through the jacket, thereby cooling or heating the reaction medium. The effectiveness of the jacket is limited by heat transfer properties which are in turn limited by mechanical design characteristics. Material of construction and wall thickness are critical design parameters for both strength and heat transfer. Unfortunately, however, heat transfer and mechanical strength are competing values in reactor design. Where the reactor wall is thick enough to meet pressure and strength requirements, heat transfer between the jacket fluid and the reaction medium is decreased.

Where the reactor wall is thinned to improve heat transfer, the structural integrity of the vessel is diminished. This trade-off has historically been the source of design efforts seeking to gain maximum heat transfer efficiency while meeting mechanical strength requirements.

In the design of reactor cooling systems, an additional concern arises when low temperature and cryogenic applications are needed. The temperature of the jacket fluid is calculated based on heat transfer requirements for a given reaction medium and reactor design. The required jacket fluid temperature is often below the freezing point of the reactor medium. As a consequence, the reactor contents can freeze along the inside of the reactor wall. This formation results in a thicker wall overall and decreased heat transfer efficiency, as well as potentially inconsistent reactor medium composition, and in some cases, destruction of some reactants or products through freezing.

An improved apparatus for controlling the temperature of a reactor during operation would allow for a thin wall and resultant increased heat transfer to the contents, without sacrificing the required mechanical properties of the reactor. Additionally, an apparatus which prevents the build-up of frozen reactor contents would maintain high heat transfer efficiency and constant reactor medium concentrations. The ideal reactor would maximize the desired properties of high mechanical strength and high heat transfer efficiency, two qualities which have historically competed.

SUMMARY OF THE INVENTION

The present invention is an insulated chemical or biological reactor (such as a fermenter) comprising a reaction vessel, an evacuated insulation shell, and a temperature controlling helical channel coil outside of the reactor but inside the evacuated shell. The channel coil is adapted to accept a circulating fluid. The particular fluid selected depends on the intended temperature control purposes, that is whether heating or cooling is desired and the degree of heating or cooling needed. The channel coil is affixed to the outside wall of the reactor in a helical configuration and the temperature controlling fluid flows spirally upward or downward around the outside of the reactor. The channel coil is shaped to have two straight, parallel sides of the coil in contact with the reactor, normal to the surface of the outside wall of the reactor. This right angle

contact between the channel coil and reactor wall increases the section modulus of the vessel wall, and thereby increases the mechanical strength of the reactor wall. The wall can thus be made thinner to promote better heat transfer across the wall. The reactor and the affixed coil are together enclosed within an evacuated jacket.

The present invention also includes a thermal probe which is immersed in the reactor contents. The thermal probe is a vertically oriented, elongated, generally cylindrical probe with an inlet and an outlet. As with the jacket, the probe may be used for heating or cooling the contents of the reaction vessel. Where heating is desired, a hot liquid or gas can be introduced into the probe through the inlet. The resultant cooler liquid or condensed vapor or liquid can be removed via the outlet. Where cooling is desired, upstream of the thermal probe inlet there is provided a phase separator to insure only a liquid stream enters the probe. The probe is placed into the top of the reactor and a liquid of desired boiling point is allowed to enter the probe while the reactor is in use. Where cooling is desired, the liquid selected would have a boiling point at or below the desired reaction temperature. The heating and boiling of the liquid introduced into the probe provides for the removal of heat from the reactor contents. For additional temperature control, the vapor produced from the boiling of the probe contents may be taken from the top of the probe and passed through the channel coil surrounding the outside of the reaction vessel.

BRIEF DESCRIPTION OF THE FIGURES

FIG. 1 is a side view of a reaction vessel with affixed channel coil of the present invention.

FIG. 2 is a sectional view of a reaction vessel with affixed channel coil in accordance with the present invention.

FIG. 3 is a sectional view of a reaction vessel with affixed channel coiled and evacuated jacket, in accordance with the present invention.

FIG. 4 is a partial sectional view of a thermal probe of the present invention.

FIG. 5 is a sectional view of the reaction vessel with affixed channel coil, evacuated jacket, and inserted thermal probe of the present invention.

FIG. 6 is a partial sectional view of the reaction vessel with affixed channel coil, evacuated jacket, two inserted thermal probes, and a mixing apparatus, according to the present invention.

FIGs. 7A and 7B are sectional views of alternative embodiments of the channel coil of the present invention.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

FIG. 1 is a view of a reaction vessel 110 with an affixed channel coil 100. The inside wall of channel coil 100 is the outside of the reaction vessel wall 120 of reaction vessel 110. The channel coil 100, before it is affixed to the reaction vessel 110, has only three outer sides, 121, 122, and 123. A fourth side of the channel coil 100 is formed by reaction vessel wall 120, being formed only upon affixation of the channel coil 100 to the reaction vessel 110. The channel coil 100 surrounds the reaction vessel 110 in a helical configuration. The configuration allows for helical and corresponding downward or upward flow, with respect to the vertical axis of the reaction vessel 110. The channel coil 100 may be constructed from any suitable material, the most likely for industrial use being carbon steel, stainless steel, Inconel (trademark for an alloy of nickel and chromium available from the Huntington Alloy Products Division of International Nickel Co. Inc. of Huntington, West Virginia), or Hastelloy C. Hastelloy is a trademark for nickel-based corrosion-resistant alloys from Union Carbide Corp. of New York, New York. Hastelloy C is a nickel-based alloy containing nickel, chromium, molybdenum, and tungsten.

FIG. 2 is a sectional view of the reaction vessel 110 with the affixed channel coil 100. FIGs. 1 and 2 show the two characteristics of channel coil 100 which combine to add mechanical strength to reaction vessel 110. The first is that the point of contact 130 is a right angle. That is, walls 121 and 123 form a right angle with wall 120. Since wall 120 is cylindrical, walls 121 and 123 must form a right angle with the axis of the cylinder at the wall. The second characteristic adding strength to reaction vessel 110

concerns the pitch at which the helical channel coil 100 is affixed to the reaction vessel wall 120. The pitch is the slope of the coil, with respect to a horizontal, radial plane which is perpendicular to the vertical axis of the reactor. A larger slope is considered a higher pitch. The channel coil 100 is affixed at a pitch less than or equal to a maximum pitch, which is that pitch beyond which the desired improvements in the reaction vessel wall 120 section modulus are no longer achieved. Exactly what this pitch is will depend on many factors including the diameter of reaction vessel 110, the thickness of reaction vessel wall 120, the material of construction of the reaction vessel 110 and the operating parameters for which the reactor is designed. As the pitch (or slope) of the coil increases, the distance between successive coils increases. Since the coil is a reinforcement to reaction vessel wall 120, as the distance between successive coils increases the degree of reinforcement decreases. At some point, the degree of reinforcement becomes too low and reaction vessel wall 120 becomes too weak for the desired function. The reinforcement required will depend upon the differential pressure between the inside and outside of reaction vessel wall 120. This is a design parameter easily calculated by one skilled in the art. Thus, the maximum pitch of channel coil 100 will depend on the designed maximum operating pressure for reaction vessel 110, among other factors.

The point of contact 130 between reaction vessel wall 120 and channel coil 110 is a right angle, and the pitch of the channel coil 100 is less than or equal to the maximum pitch. These two factors combine to increase the section modulus of the reaction vessel 110. This resultant increase in the section modulus, due to the channel coil 100, allows the reaction vessel wall 120 be thinner than that which would otherwise be required when the channel coil 100 is not affixed according to the present invention. Because the reaction vessel wall 120 may be thinner than that which would be required without channel coil 100, improved heat transfer efficiency is achieved. A thinner reaction vessel wall increases the overall heat transfer coefficient across the reaction vessel wall because the thermal resistance resulting from the thermal conductivity of the reaction vessel wall is reduced.

The channel coil 100 may be additionally insulated with insulation 700 attached directly to the three outer sides, 121, 122, and 123, of the coil 100 as shown in

Figs. 7A or 7B. Alternatively, insulation 700 may be wrapped around channel coil 100 and reaction vessel 110 as shown in figure 2, before placement of evacuated shell 300. Insulation 700 may be any suitable material. Reflective multilayer insulation, fiberglass cloth, or aluminum foil are preferred. These materials may also be used alone or in combination with one another.

FIG. 3 is a sectional view of reaction vessel 110 with the affixed channel coil 100, and an evacuated shell 300. The evacuated shell 300 completely encloses reaction vessel 110 and channel coil 100, with the exception of related piping and utilities which penetrate the evacuated shell 300. The placement of the evacuated shell 300 around the apparatus as described above allows for additional insulation of reaction vessel 110 and channel coil 100 from the ambient air. Insulation from the ambient air results in decreased heat transfer through both the reaction vessel wall 120 and the channel coil walls 121, 122, and 123. The utilization of evacuated shell 300 results in greater temperature control of the reaction vessel contents. The evacuated shell may be constructed from any suitable material, including carbon steel, stainless steel, Inconel, or Hastelloy C. Further, evacuated shell 300 can also include reflective material on the inner or outer surface thereof to reduce radiant heat transfer.

FIG. 4 is a partial sectional view of a thermal probe in accordance with the present invention. In the exemplary embodiment, a thermal probe 400 is used where there exists a need to cool the reaction vessel contents. However, such probes can also be used where heating of the contents of reaction vessel 110 is needed. The thermal probe 400 is inserted into the reaction vessel contents as shown in FIG. 5. For cooling, a liquid is introduced into the thermal probe 400 through an inlet pipe 410. As previously discussed, the liquid is selected primarily because of its boiling point, providing, of course, other factors do not prevent its use, such as availability, cost, reactivity, toxicity, etc. A liquid having a boiling point lower than that of the reaction vessel contents will boil when heat is absorbed from the reaction vessel contents. Fluids which may be used for cooling or heating in the present invention include, but are not limited to nitrogen, brine, steam, chilled water, carbon dioxide, and hot water. Other fluids may also be used depending on the particular needs of the reaction for which the reactor is designed.

The ideal temperature (or range of temperatures) of the reaction vessel contents can be determined from the chemistry of the reaction. This temperature, along with the physical characteristics of the thermal probe (dimensions, material of construction, number of probes, etc.) and relevant heat transfer equations, are combined to give rise to a required amount of heat transfer which must occur across the wall of the thermal probe in order to maintain the reactor contents at the desired temperature. From this required value of heat transfer, a fluid is selected such that the latent heat of vaporization plus any sensible heat transfer occurring from any rise in temperature of the fluid to its boiling point, will give the desired total heat transfer. It should be noted that a fluid with precisely the right characteristics does not have to exist for accurate control of the temperature. Controlling the flow rate of the fluid into the probes will allow for fine tuning the heat transfer and corresponding temperature of the reactor contents. Further, controlling the pressure of the liquid could help alter its boiling point and fine tune the cooling power and range of the liquid. The selected fluid need only fall within a range of necessary heat transfer requirements. Where heating is desired, a hot gas, such as hot nitrogen, is introduced into channel coil 100 as well as probe 400. This hot gas then heats the contents of reaction vessel 110.

For instance, if a higher rate of cooling is desired, then fluid flow into the probe 400 can be increased. This will raise the level 448 of liquid 450 in the probe 400. This in turn will expose a greater surface area of liquid 450 to wall 449 of probe 400, thus allowing greater heat transfer from the reaction vessel contents through wall 449 into liquid 450.

Alternatively, the thermal probes could be used in one of several different heating and cooling schemes. The probes may be used to gain only sensible heat. The probes could also be utilized with a liquid having a boiling point higher than the desired temperature of the reactor contents. Cooling or warming liquid could be passed through the probes. Additionally, a gas may be passed through the probes. Any fluid that provides the necessary heat transfer properties could be used in the probes for effective temperature control of the reaction vessel contents. In these cases, the probes act as simple heat exchangers.

Probes can be inserted from the top of the reactor or from the bottom of the reactor. In the preferred embodiment, probes have been inserted from the top as a matter of convenience and tradition.

FIG. 4 also shows a sintered, porous metal phase separator or "snubber" 411 placed at the end of inlet pipe 410. The snubber 411 disturbs the flow of the liquid into the probe, just as a kitchen faucet nozzle aerates the water flow into a sink, thereby minimizing splashing. Snubber 411 also serves to disengage and allow the phases to separate.

FIG. 4 also shows a means for the gas formed from the boiling liquid to escape. An annular space 420 surrounds the inlet pipe 410. Annular space 420 comprises the same atmosphere as that above the liquid level in the thermal probe 400. As liquid flows into the thermal probe 400 through inlet pipe 410, resultant vapor or gas is pushed upward and out of the thermal probe 400 through exit 430. The exiting gas may then be utilized in various ways. If environmentally safe gas is used, it may be exhausted to the atmosphere by venting it, although this is likely not cost effective. The gas may be recovered by piping it to a condenser, or used at another site where the particular vapor or gas is needed. Finally, the exiting gas may be transported, through vacuum jacketed or otherwise insulated pipe, to the channel coil 100 for further cooling of the reactor contents.

FIG. 4 also shows a means for detecting the level of liquid in the thermal probe 400. A dual leg dip tube 440 is inserted into the thermal probe 400. The top opening 445 of the dip tube 440 is near the top of the thermal probe 400, and the bottom opening 447 of the dip tube 440 is near the bottom of the thermal probe 400. The level of liquid 450 in the thermal probe 400 is maintained below the top opening 445 and above the bottom opening 447 of the dip tube. The pressure differential is detected as the pressure of the head of liquid in the dip tube. The pressure at the top opening 445 is the pressure of the gas above the liquid 450. The pressure at the bottom opening 447 is the pressure of the gas above the liquid 450 plus the pressure caused by the weight of the liquid 450 which is above the bottom opening 447. The pressure created by the weight of liquid 450 above the bottom opening 447 can be found by subtracting the value of the

pressure at the top opening 445 from the value of the pressure at the bottom opening 447. This pressure can be used (in conjunction with the density of the liquid) to calculate the height of liquid above the bottom opening 447.

FIG. 5 shows the present invention including reaction vessel 110, channel coil 100, and one thermal probe 400. It would be apparent to one of ordinary skill in the art that multiple thermal probes could be used to increase the overall rate of heat transfer between the reactor contents and thermal probe contents. An additional advantage to utilizing multiple thermal probes is seen where the reactor contents are agitated with a mixing blade apparatus. FIG. 6 shows an embodiment where multiple probes are used in conjunction with an agitator. In such a case, the thermal probes 400 must be arranged outside the radius of mixing blades 600. In such a configuration, the thermal probes also act as mixing baffles.

Where the reactor is agitated as shown in FIG. 6, formation of frozen reactor contents on the outside surface of the thermal probes is minimized by placing the probes in or near the streamlines corresponding to maximum free stream velocity. By placing the probes in these high velocity streamlines, turbulent flow around the probes is maximized. By maximizing turbulent flow immediately adjacent to the probes, the thickness of the laminar thin film at the surface of the probe is minimized. Minimizing the thickness of this film is important in preventing material from solidifying on the surface of the probe.

The correlation between high turbulence and decreased ice (or solid) formation is due to the fact that heat transfer through a laminar layer is largely conduction controlled, but heat transfer through a turbulent fluid is largely convection controlled. Convective heat transfer takes place because a fluid is in motion and eddies within the fluid effectively carry heat throughout the fluid. This is very efficient heat transfer. Conductive heat transfer, however, is due to molecular interaction between the molecules comprising the medium through which the heat passes. This type of heat transfer is much less efficient than convective heat transfer. Where heat transfer is convection controlled, it occurs much more quickly than for the same fluid, not moving, where conduction is the only source of heat transfer. Moreover, when a fluid has turbulent flow characteristics,

heat transfer is much quicker than where the same fluid is not moving (and other pertinent factors are the same). So where the laminar, non-moving, fluid film thickness is minimized, more heat is transferred through it in a given time period and the formation of ice is subsequently slowed or prevented. Where the laminar layer is thick, heat transfer is limited, and the layer freezes more quickly than where the layer is thinner. The above mentioned probe placement provides for an overall heat transfer coefficient that is largely convection-controlled, corresponding to fully developed turbulent flow. This maximizes overall heat transfer and prevents build-up of frozen reactor contents on the probe surface.

FIGs. 7A and 7B show additional embodiments of the shape of channel coil 100. The outside wall 122 of the channel coil 100 may be of nearly any shape. It is critical, however, that the walls 121 and 122 are both normal to the outside reaction vessel wall 120. In this configuration, channel coil 100 supports and strengthens reaction vessel wall 120, allowing use of a thinner wall and greater heat transfer.

Although the present invention has been described with reference to exemplary embodiments, it is not limited thereto. Rather, the appended claims should be construed to include other variants and embodiments of the invention which may be made by those of ordinary skill in the art without departing from the true spirit and scope of the present invention.

What is Claimed:

1. An insulated chemical reactor comprising:

a reaction vessel having inner and outer surfaces;

an evacuated insulation shell; and,

a temperature controlling helical channel coil outside of said reaction vessel and within said shell,

wherein said reaction vessel forms one wall of said channel coil.

2. A reactor as claimed in claim 1, wherein said temperature controlling helical channel coil comprises a rectangular C-shaped channel affixed to said outer surface of the reaction vessel, said channel coil having at least one wall in contact with said reaction vessel normal to the surface of said reaction vessel.

3. A reactor as claimed in claim 2, wherein said channel coil comprises two flat, parallel walls, each in contact with said reaction vessel normal to the surface of said reaction vessel coil.

4. A reactor as claimed in claim 1, further including insulating material affixed to said channel coil.

5. A reactor as claimed in claim 1, wherein said fluid is selected from the group consisting of nitrogen, brine, steam, chilled water, carbon dioxide, and hot water.

6. An apparatus for cooling contents of a reaction vessel having a top and a bottom, by allowing a liquid to boil inside a probe immersed in the reactor contents, to produce gas inside the probe, comprising:

a vertically oriented, elongated cylindrical probe having a top and a bottom, said probe immersed in the contents;

means for introducing said liquid into the top of said probe to a predetermined level;

means for removing said gas from said probe; and,

means for controlling the level of liquid in said probe.

7. An apparatus as claimed in claim 6, wherein said means for introducing said liquid includes a phase separator.

8. An apparatus as claimed in claim 6, wherein said means for introducing said liquid comprises an inlet line extending through the top of said probe, and extending coaxially through a portion of said probe.

9. An apparatus as claimed in claim 6, wherein said means for removing said gas comprises an outlet line in fluid communication with the top of said probe.

10. An apparatus as claimed in claim 6, wherein a multiplicity of probes are immersed in the contents.

11. An apparatus as claimed in claim 6, wherein said probe is inserted into the reaction vessel through the top of the reaction vessel.

12. An apparatus as claimed in claim 6, wherein said probe is inserted into the reaction vessel through the bottom of the reaction vessel.

13. An apparatus for supplying gas to a temperature controlling helical channel coil, comprising:

a thermal probe, immersed in the reactor contents, wherein a liquid is boiled;

means for supplying gas discharged from said probe;

means for monitoring the flow of said gas into the helical channel coil; and,

means for controlling the flow of gas into the helical channel coil.

14. An apparatus as claimed in claim 13, wherein said liquid is selected from the group consisting of nitrogen, brine, steam, chilled water, carbon dioxide, and hot water.

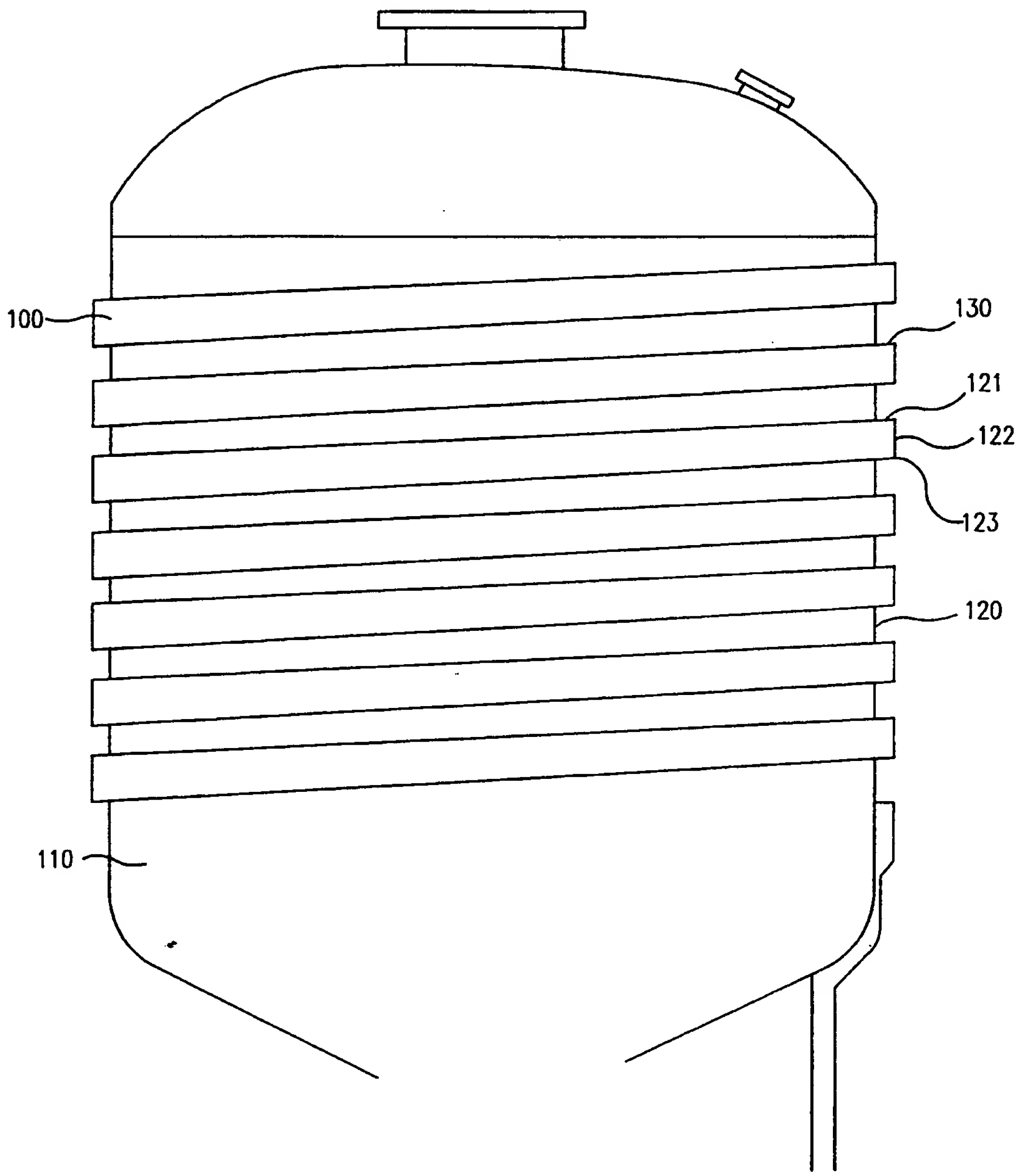

FIG. 1

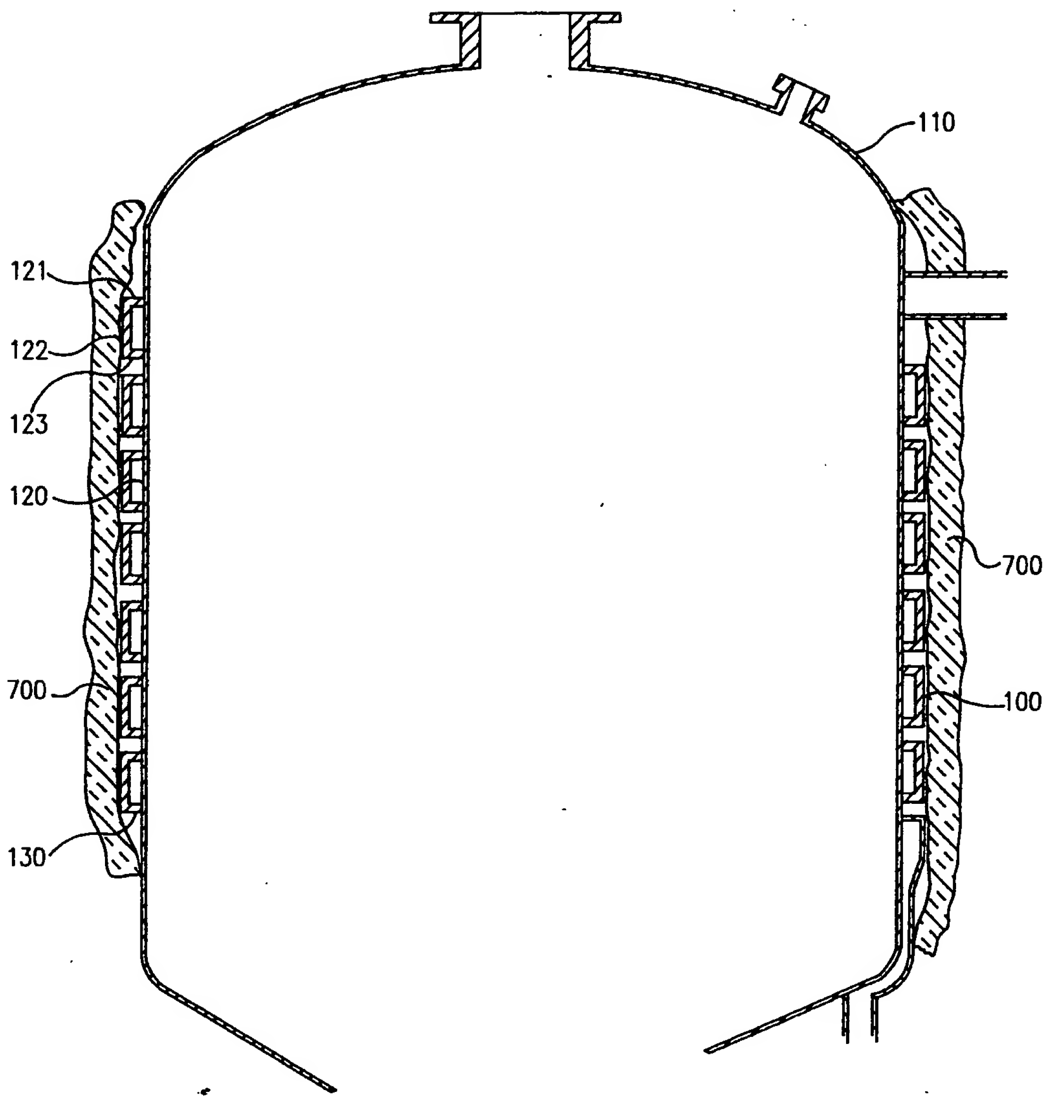

FIG. 2

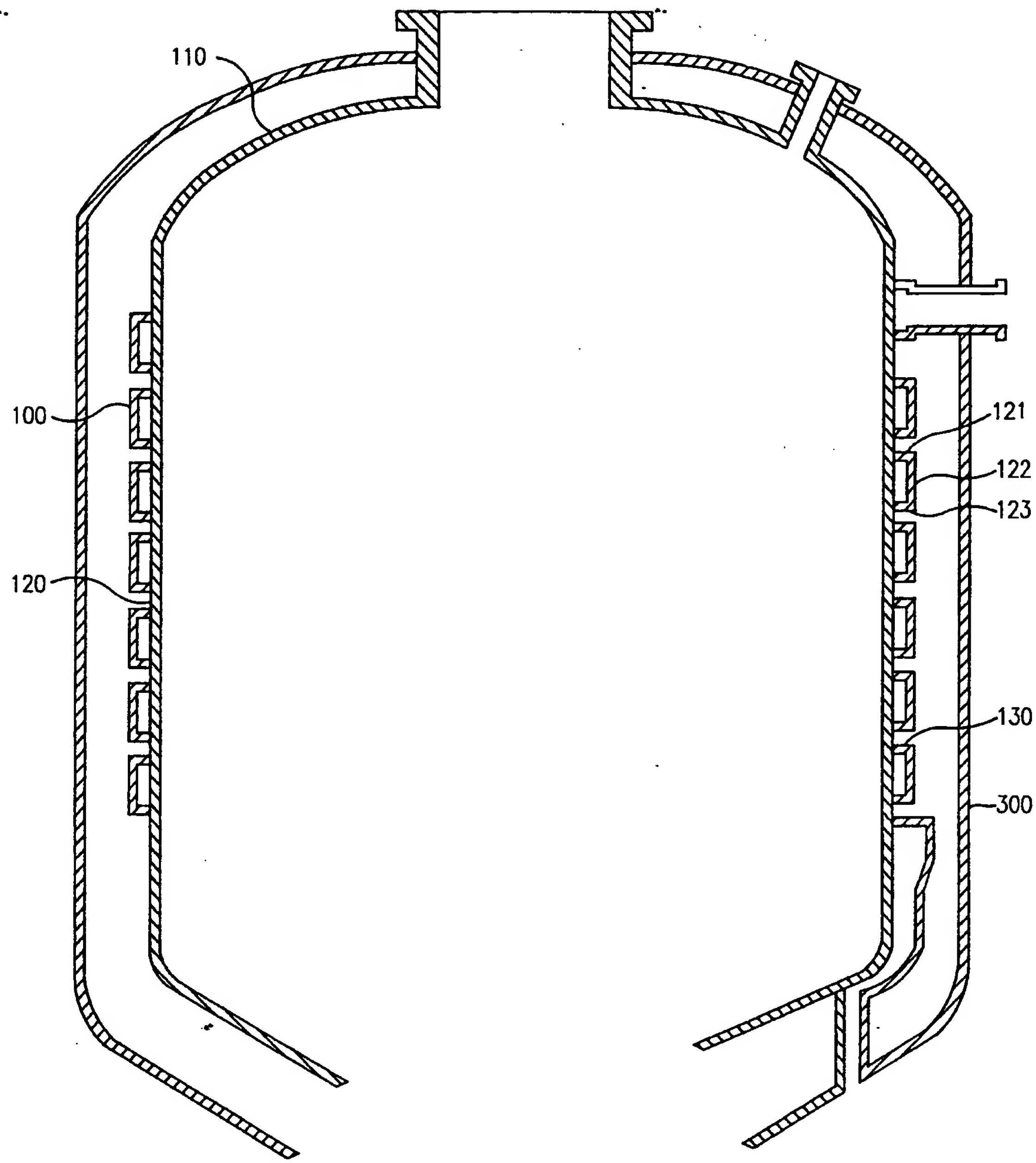

FIG. 3

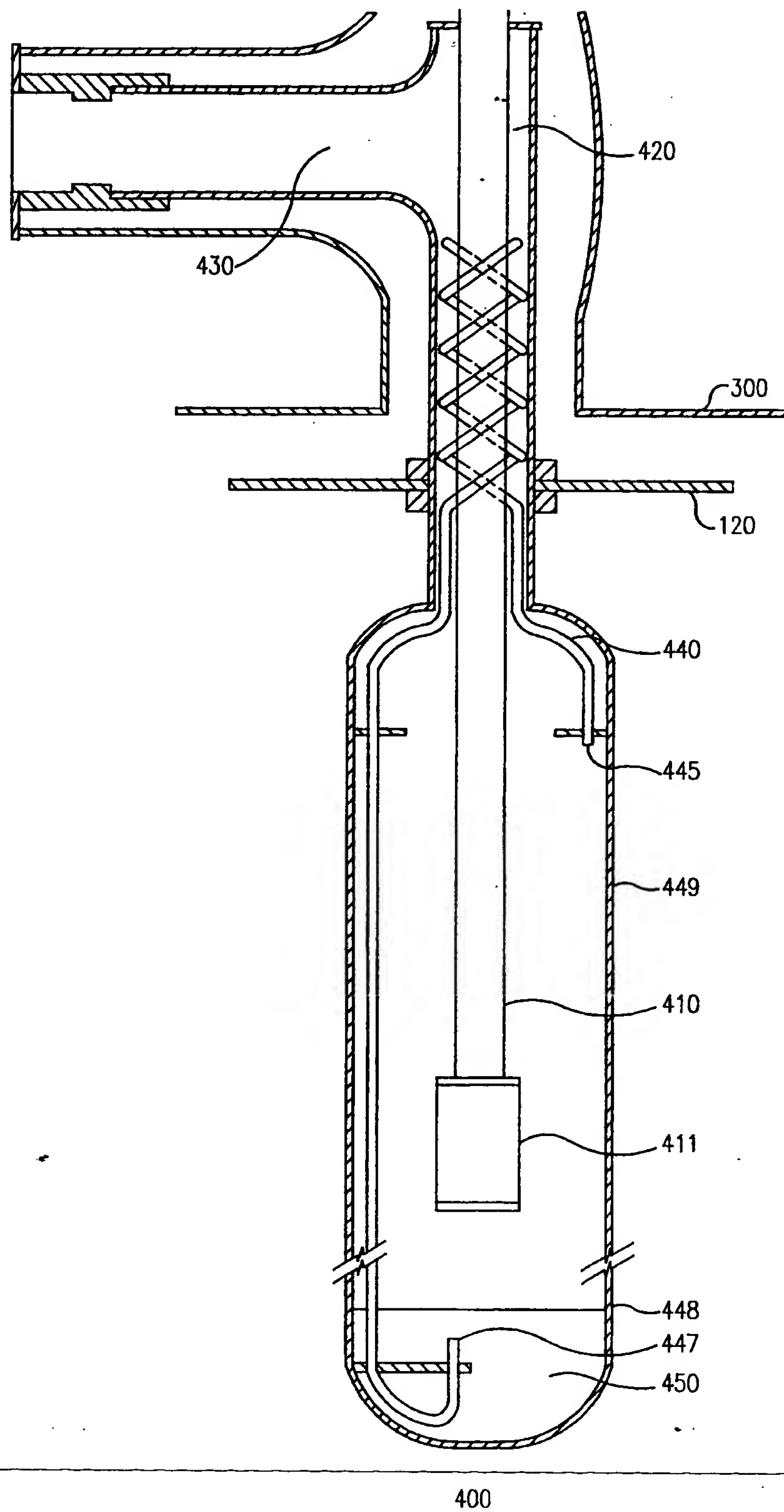

FIG. 4

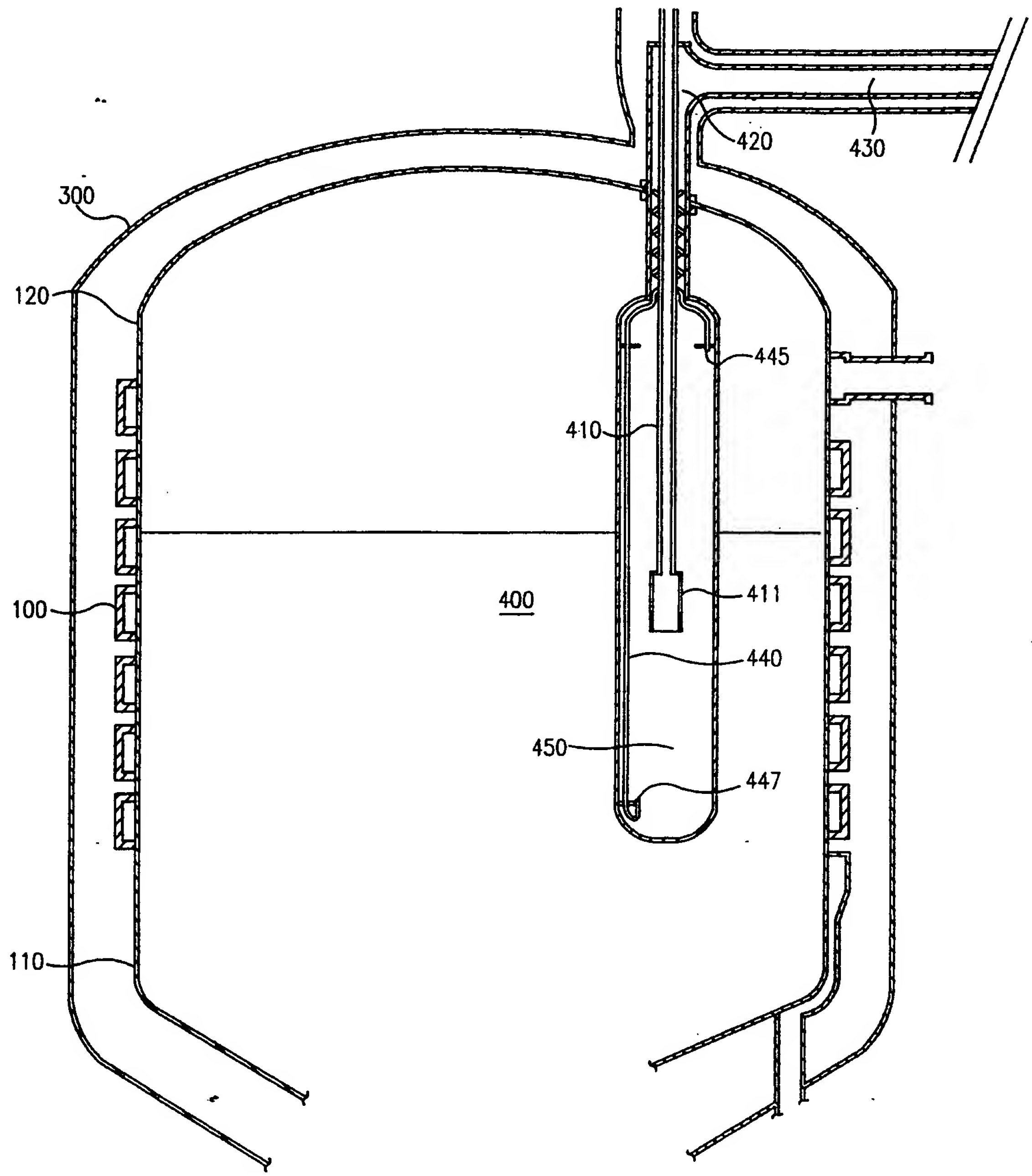

FIG. 5

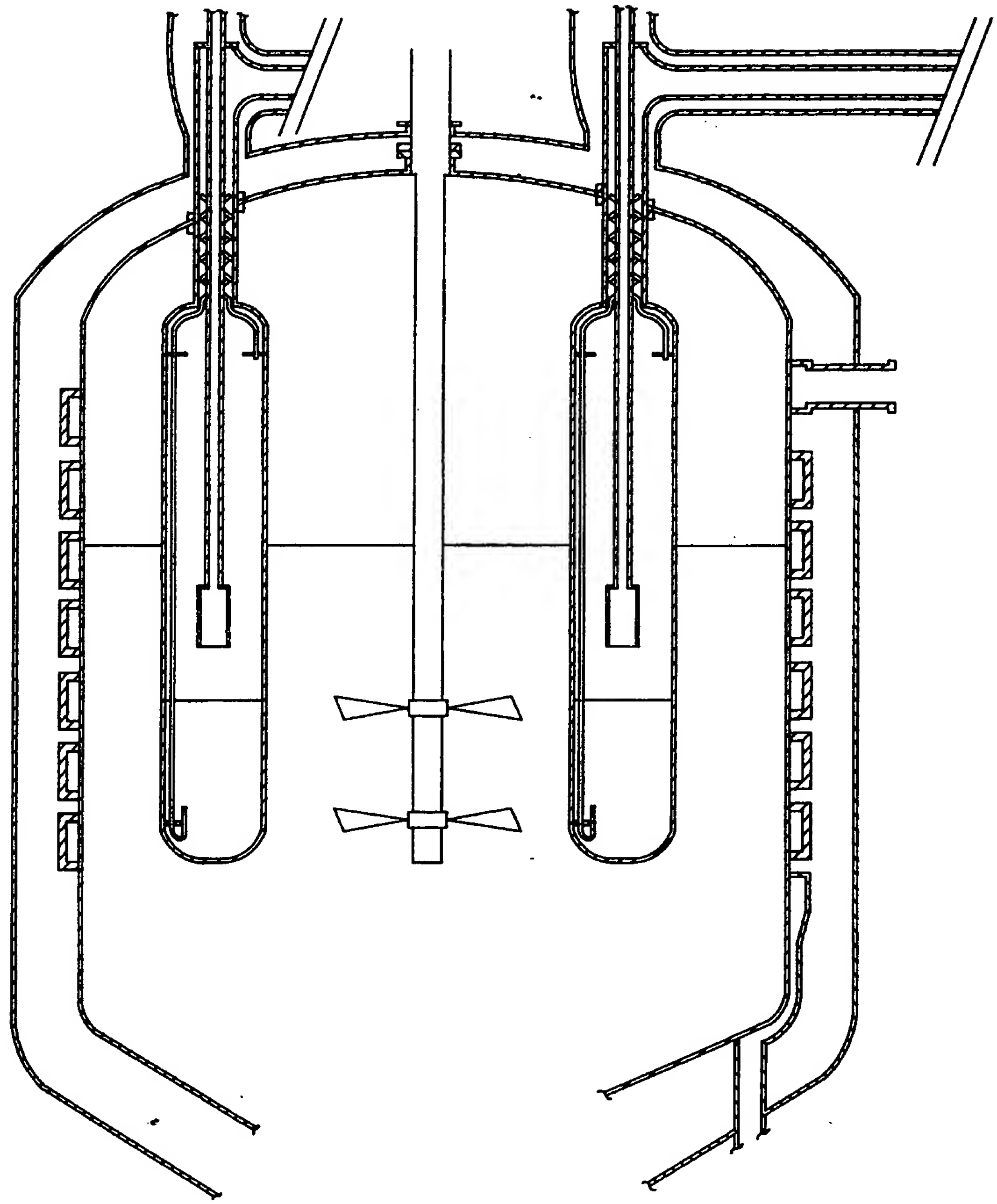

FIG. 6

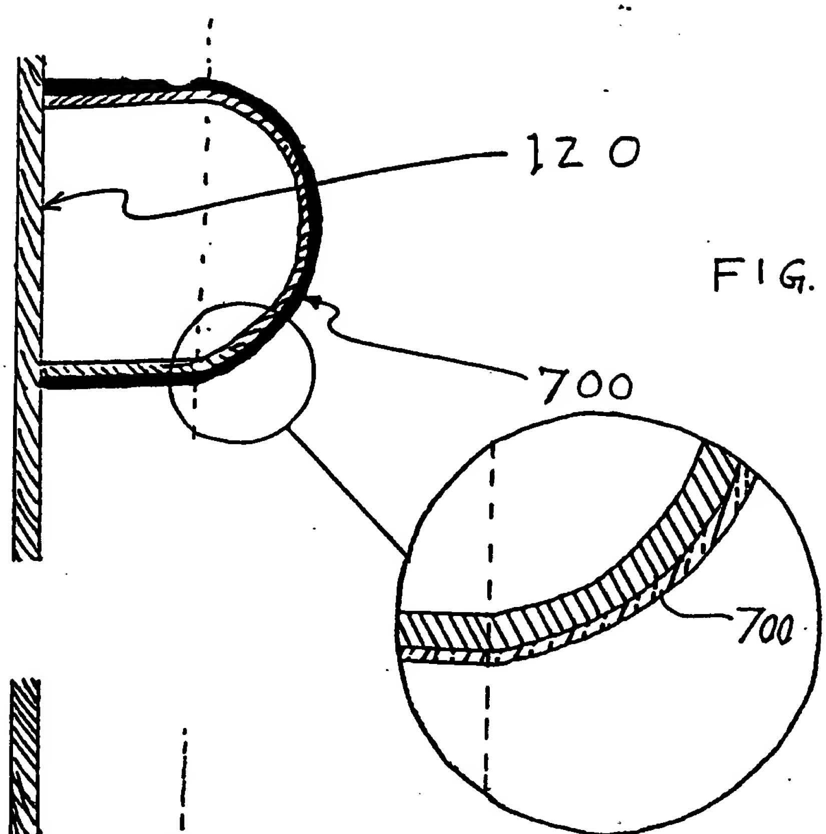

FIG. 7A

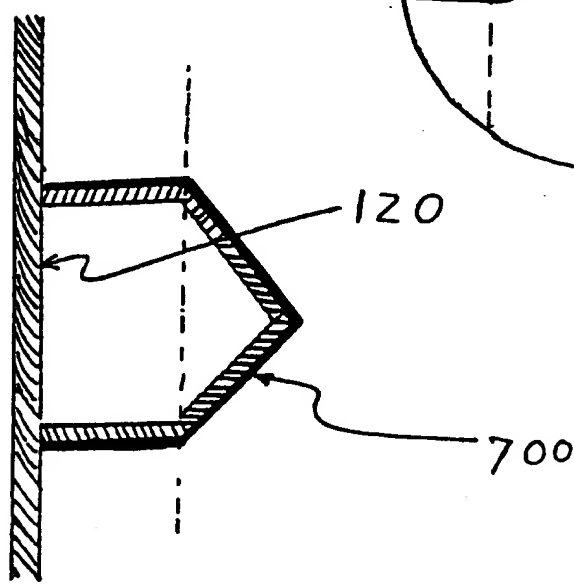

FIG. 7B

RECEIVED PATENT COOPERATION TREATY

MAR 24 1999

From the INTERNATIONAL SEARCHING AUTHORITY

~~RATNER & PRESTIA~~

Art. 19 5/19/9
IOS in com. US
PCT Appn. 6/19/99.

To:

RATNER & PRESTIA
Attn. SIMMONS, J.
P.O.Box 980
Valley Forge
Pennsylvania 19482-0980
UNITED STATES OF AMERICA

NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL SEARCH REPORT
OR THE DECLARATION

(PCT Rule 44.1)

ECG - 019

Comments (
Abstract 4/19/

	Date of mailing (day/month/year) 19/03/1999
Applicant's or agent's file reference ECG-019	**FOR FURTHER ACTION** See paragraphs 1 and 4 below
International application No. PCT/US 98/12102	International filing date (day/month/year) 15/06/1998
Applicant ARENCIBIA ASSOCIATES, INC.	

1. [X] The applicant is hereby notified that the International Search Report has been established and is transmitted herewith.

Filing of amendments and statement under Article 19:
The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):

When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.

Where? Directly to the International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland
Fascimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. [ ] The applicant is hereby notified that no International Search Report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. [ ] **With regard to the protest** against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

   [ ] the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.

   [ ] no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after **18 months** from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90*bis*.1 and 90*bis*.3, respectively, before the completion of the technical preparations for international publication.

Within **19 months** from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).

Within **20 months** from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the International Searching Authority	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Trudy Thoen-de Jong

Form PCT/ISA/220 (July 1998)

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions, respectively.

INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

When? Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

How? Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped),whether

(i) the claim is unchanged;

(ii) the claim is cancelled;

(iii) the claim is new;

(iv) the claim replaces one or more claims as filed;

(v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]: "Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."

2. [Where originally there were 15 claims and after amendment of all claims there are 11]: "Claims 1 to 15 replaced by amended claims 1 to 11."

3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]: "Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or "Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."

4. [Where various kinds of amendments are made]: "Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

"Statement under article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments and any accompanying statement, under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the time of filing the amendments (and any statement) with the International Bureau, also file with the International Preliminary Examining Authority a copy of such amendments (and of any statement) and, where required, a translation of such amendments for the procedure before that Authority (see Rules 55.3(a) and 62.2, first sentence). For further information, see the Notes to the demand form (PCT/IPEA/401).

Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference ECG-019	**FOR FURTHER ACTION** see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/US 98/12102	International filing date *(day/month/year)* 15/06/1998	(Earliest) Priority Date *(day/month/year)* 18/06/1997
Applicant ARENCIBIA ASSOCIATES, INC.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 6 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

   a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

   [ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

   b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

   [ ] contained in the international application in written form.
   [ ] filed together with the international application in computer readable form.
   [ ] furnished subsequently to this Authority in written form.
   [ ] furnished subsequently to this Authority in computer readble form.
   [ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
   [ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).

3. [X] **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

   [X] the text is approved as submitted by the applicant.
   [ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

   [ ] the text is approved as submitted by the applicant.
   [X] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. 5

   [ ] as suggested by the applicant.
   [ ] None of the figures.
   [ ] because the applicant failed to suggest a figure.
   [X] because this figure better characterizes the invention.

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

SEE ADDITIONAL SHEET

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☒ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

GROUP II: CLAIMS 6-12 AND
GROUP III: CLAIMS 13,14

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1-5

   The subject- matter of the first invention deals with an insulated chemical reactor comprising a reaction vessel having inner and outer surfaces, an evacuated insulation shell, a temperature controlling helical channel coil outside of said reaction vessel and within said shell, the reaction vessel forming one wall with the channel coil. The helical coil is further disclosed in the sub-claims 2-4 (shape, position relative to the outer surface of the reaction vessel, insulation material affixed to the coil). A variety of fluids used as cooling or heating media is finally given in sub-claim 5.

2. Claims: 6-12

   The subject- matter of the second invention deals with an apparatus for cooling the contents of a reaction vessel. This apparatus is immersed in the reactor contents and comprises a vertically orientated, elongated cylindrical probe having a top and a bottom, means for introducing a liquid into the top of the probe to a predetermined level and means for controlling the level of liquid inside the probe. The liquid is allowed to boil inside the probe thereby producing a gas. The gas is removed from the probe by suitable means.
   The means for introducing the liquid into the probe are described in sub- claims 7 and 8.
   The means for removing the gas from the probe are disclosed in sub- claim 9.
   The position of the probe in the reaction vessel and the number of probes inserted in the vessel are disclosed in sub- claims 10-12.

3. Claims: 13,14

   The subject- matter of the third invention deals with an apparatus for supplying gas to a temperature controlling helical channel coil comprising a probe filled with liquid immersed in the reactor contents, means for supplying gas discharged from the probe, means for monitoring the flow of gas in the helical channel coil and means for controlling the flow of gas in the channel coil.
   In a further sub- claim the liquid is said to be selected from a given variety of substances.

Box III TEXT OF THE ABSTRACT (Continuation of it m 5 of the first sheet)

```
Line 2 : after "apparatus" add (110)
         after "vessel" add (110)
line 3 : after "probes" add (400)
line 5 : after "coils" add (100)
line 6 : after "coils" add (100)
         after "vessel" add (110)
line 8 : after "coils" add (100)
         after "vessel" add (110)
line 11: after "shell" add (300)
```

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B01J19/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 B01J F17C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	GB 871 752 A (CIBA LIMITED) 28 July 1961 see claims 1-6 see figures 1-3 ---	1,2,4
Y	US 5 375 423 A (DELATTE DANIEL) 27 December 1994 see the whole document ---	1,2,4
A	EP 0 423 944 A (TOSOH CORP) 24 April 1991 see page 1, line 29 - line 32 see page 2, line 21 - line 51 ---	1,5,6,13
A	EP 0 659 475 A (SUMITOMO HEAVY INDUSTRIES) 28 June 1995 see abstract; figures 1,2 --- -/--	1,2

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
3 March 1999	19.03.99

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Vlassis, M

3

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 20 05 145 A (JACOB AND KORVES GMBH) 19 August 1971 see page 3, paragraph 1 - page 4, paragraph 1 see page 5, paragraph 1 - page 7, paragraph 1 see figures 1-4 ---	1-3
X	GB 467 564 A (ROBINSON BINDLEY PROCESSES LTD) 18 June 1937 see page 3, left-hand column, line 23 - page 4, right-hand column, line 83 see figure 1 ---	6,8,9,12
A	DE 19 58 261 A (THOMSON-CSF) 4 June 1970 see page 2, paragraph 5 - page 4, paragraph 3 see figures 1,2 ---	6,10-12
A	US 2 537 472 A (MASSIOT L,) 9 January 1951 see claims 1-6; figures 1-5 ---	6,8-12
Y A	US 4 029 143 A (GOEBEL PAUL) 14 June 1977 see column 2, line 5 - line 55 see column 3, line 60 - column 4, line 5 see column 4, line 46 - line 65 see figures 4,5 ---	13,14 6,11
Y	DATABASE WPI Section Ch, Week 9028 Derwent Publications Ltd., London, GB; Class J04, AN 90-215112 XP002095502 & SU 1 511 737 A (AKRIM CHEM. PHARM., LENGO LENSOVET TECH.), 30 September 1989 see abstract ---	13,14
A	PATENT ABSTRACTS OF JAPAN vol. 017, no. 102 (C-1031), 2 March 1993 & JP 04 293538 A (TOSOH CORP), 19 October 1992 see abstract -----	6,13

3

Patent document cited in search report		Publication date	Patent family member(s)			Publication date
GB 871752	A		CH	353341	A	
			DE	1094718	B	
			FR	1205978	A	05-02-1960
US 5375423	A	27-12-1994	FR	2697074	A	22-04-1994
			DE	4334990	A	28-04-1994
			GB	2271839	A,B	27-04-1994
EP 0423944	A	24-04-1991	JP	3115303	A	16-05-1991
			US	5131232	A	21-07-1992
EP 0659475	A	28-06-1995	JP	7185314	A	25-07-1995
			CN	1108965	A	27-09-1995
			US	5667758	A	16-09-1997
DE 2005145	A	19-08-1971	NONE			
GB 467564	A		NONE			
DE 1958261	A	04-06-1970	AT	302249	B	15-09-1972
			FR	1595711	A	15-06-1970
			GB	1269250	A	06-04-1972
			US	3646320	A	29-02-1972
US 2537472	A	09-01-1951	NONE			
US 4029143	A	14-06-1977	DE	2441302	A	11-03-1976
			DE	2504926	A	19-08-1976
			AU	8435875	A	03-03-1977
			BE	832920	A	01-03-1976
			BR	7505523	A	03-08-1976
			DK	387875	A	01-03-1976
			FR	2283151	A	26-03-1976
			GB	1514794	A	21-06-1978
			IN	144018	A	11-03-1978
			JP	51050386	A	01-05-1976
			LU	73260	A	15-04-1977
			NL	7509961	A	02-03-1976
			ZA	7505506	A	25-08-1976